Supporting Information

Spontaneous Self-welding of Silver Nanowire Networks

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Experimental Section

Silver nanowire deposition: Aqueous 1 wt % of Ag NWs suspension was purchased from N&B Company, Korea. Diameter and length of the NWs were 50 nm and 20 μ m, respectively. We conducted all experiments at room temperature. The electric field-assisted spray nozzle developed to atomize the liquid into nanoscale like the conventional electrospray. It could overcome the drawback of the limited flow rate. The nozzle structure consisted of a liquid core nozzle and a sheath nozzle for gas flow (Figure. S1). A ceramic nozzle tip with an outer diameter of 250 μ m and inner diameter of 150 μ m was equipped at the end of the liquid core nozzle. The outlet of the sheath nozzle was 2 mm diameter. Electrical potential was produced by the high voltage module. Compressed air pressures were supplied to the nozzle perpendicular to the direction of the liquid ejection through two asymmetric holes. The coating system consisted of a nozzle, x-y motorized substrate, high voltage controller, vision module, electrical current measurement module (Figure. S1). High voltage could be supplied to a nozzle and grounded 50 cm × 50 cm² substrate was used to fix the 30 cm × 30 cm² PET film. A distance between nozzle and substrate was 4 cm.

Numerical simulation

The COMSOL Multiphysics FEMLAB program was used for the temperature calculations of the Ag NWs junction. An unstructured mesh with 12195087 elements was used for the simulation. A governing equation for electric currents was defined by $E = -\nabla V$. The governing equation for heat transfer was defined by $\rho C_p u \cdot \nabla T = \nabla \cdot (k \nabla T) + Q$. The contact conductivity was estimated by Cooper-Mikic-Yovanovich correlation. The 'contact conductivity' parameter was extracted by solving $h_c = 1.25\sigma_{contact} \frac{m_{asp}}{\sigma_{asp}} \left(\frac{P}{H_c}\right)^{0.95}$, where $\sigma_{contact}$ is the surface area of the contact area, m_{asp} is the surface roughness of the asperities average slope, σ_{asp} is the surface roughness of the asperities' average height, P is the contact pressure, and H_c is hardness. Temperature distributions were achieved from the simulation and the maximum temperature at the contact area was evaluated. The contact pressure (2 kPa) was assumed to be kinetic energy of the droplet impacted on the substrate.

Characterization

The spray distributions were captured using a Photron Ultima APX high speed camera at 250 frames per second. A continuous laser with a cylindrical quartz glass plate for the laser beam was equipped. The spray fluctuations were evaluated by the 100 frames using home-made software. The thin films with the Ag NWs network were characterized using a JEOL JEM-7600F Field Emission Scanning Electron microscope and SII NANONAVI E-sweep Atomic Force microscopy. The sheet resistances and the transmittance were measured using an EDTM 4 point probe and Agilent Carry 5000 UV-VIS-NIR system, respectively. The electrical and optical performances were evaluated on 5 different points.

Measurement of the droplet velocity: The spraying was monitored by a Photron RS-APX high speed camera whose frame-rate was 1/6000 s. A continuous laser which ranges from 0 to 4 W was used with the cylindrical quartz glass plate for the laser-beam. The images of 200 frames were analysed by particle image velocimetry (Davis Inc.) to evaluate the velocity components and the trajectory of droplets generated by spray.

Measurement of electrical current: For the simulation of the joule heating case and current density, the currents were measured using a Keithley 6485 picoammeter. The current was measured for 8 s at 64 ms intervals. Before depositing the Ag NWs solutions, a measuring probe was connected to the PET. Because the sprayed droplets had fast evaporation and immediately made the thin film electrode, we could measure the current of the Ag NWs film on the PET.



Figure S1. Experiment setup for electric field-assisted spray and length distribution of silver

nanowires







Figure S3. (a) Pixel analysis of spray images with and without electric field. The spray distribution could be improved more uniformly along the transverse direction with the electric

field. The grey scale was used for analyzing the brightness of spray distributions. The brightness (green) were evaluated at 20 mm away from the nozzle outlet. Under the 20 kPa of gas pressure, image of spray distributions with (b) 0 kV and (c) 4 kV. (d) Standard deviations of spray fluctuations with and without electric field. Standard deviations were calculated with 100 frames of captured images using a high speed camera. As the electric field was imposed on the nozzle (blue), the standard deviations decreased at the edge of the spray distribution. On the other hand, the fluctuation widens when the electric field strength is too strong (red) because the droplets affected by the vortex at the boundary area move towards the substrate through the electric field streamlines



Figure S4. Effect of air pressure on the Ag NW film electrical and optical performances without the electric field. As pressure increases to over 50 kPa, much spray might be lost causing high sheet resistance and fluctuation of the spray made non-uniform film quality. Therefore, one should use low pressure so that we used the 20 kPa for the AgNWs coating.



Figure S5. Thickness of Ag NWs: (a) Junction of self-welded NWs (A: 73.65 nm), (b) Single NW (B: 49.66 nm and C: 55.42 nm), (c) Junction of air sprayed NWs (D: 93 nm) and (d) Single NW (E: 49.24 nm)



Figure S6. (a) Streamlines of current for nanowires. When the current flows from the top nanowire to the bottom nanowire, it is concentrated at the contact area. (b) Mesh for simulation consisted of 12195087 elements. (c) Temperature result of 7 nm overlap thickness.
(d) Temperature result of overlap thickness of 5 nm. (e) Temperature result of 3 nm overlap thickness. (f) Temperature result of 1 nm overlap thickness.



Figure S7. (a) Sheet resistance obtained on the PET (30 cm \times 30 cm2) substrate at the 5 different points using a 4 point probe. The average of the sheet resistance and the standard deviation was 59.84 Ω sq⁻¹ and 0.88, respectively. (b) Evaluation of uniformity for sheet resistance at the 5 different points. The operating air pressure of 130 kPa (black) caused significant deviation of sheet resistance. However, the reduced operating air pressure of 20 kPa (red) resulted in uniform and also decreased sheet resistance. Moreover, as the electric field was imposed on the nozzle, we could achieve highly uniform results. It is expected that

the operating pressure and the electric field can solve the material loss problem as well as the

uniformity of the thin film.